

Case EL/2-22714/A/PCT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF  
ROMAN LENZ ET AL.  
SERIAL NO. 10/521,651  
FILED: January 13, 2005

Group Art Unit: 1714  
Examiner: KRIELLION A. SANDERS

FOR: HIGH-MOLECULAR-WEIGHT POLYMERIC MATERIAL  
COMPRISING DIKETOPYRROLOPYRROLE PIGMENTS

Commissioner for Patents  
Washington, D.C. 20231

DECLARATION UNDER 37 CFR 1.132

I, Roman Lenz, a citizen of Switzerland, residing in 4410 Liestal, Switzerland, hereby declare:

That the University of Basel, Switzerland, awarded me the degree of Doctor of Sciences (chemistry) in 1996;

That I have 9 years of experience in the field of organic dyestuffs and pigments, especially in the synthesis and screening of organic pigments;

That for 9 years I have been in charge of a lab team dealing with polycyclic pigments, especially of the DPP type, for the coloration of high molecular weight organic materials within CIBA-SC, Basle, Switzerland;

That I am the inventor or co-inventor of a number of patents, including US Patents in fields closely related to that of the above-identified application;

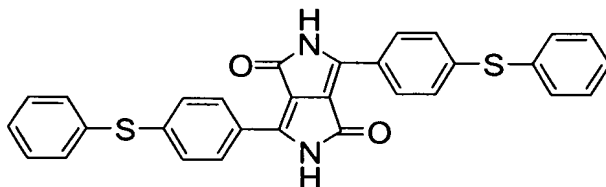
That I am the main inventor of the instant application 10/521,651, the contents of which I have read carefully;

That I have also read US Patents No. 4,579,949 (Rochat et al.), 5,738,719 (Wallquist et al.) and the PCT patent application WO-02/10288 (de Keyzer), and that I am very familiar with the subject matter thereof;

That the experiments described in the following have been carried out by me or under my supervision and the evaluation of the test results has been done by myself.

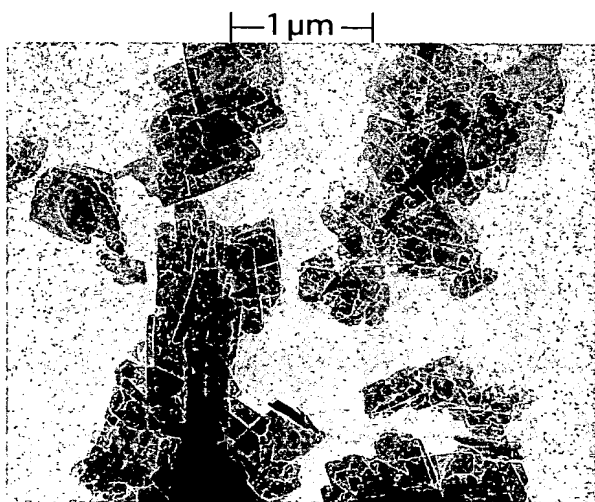
## EXPERIMENTAL

Comparative sample A was made according to the method of example 48 of '949, with the only difference that a molar equivalent amount of p-phenylthio-benzonitrile was used instead of benzonitrile. The pigment of following formula was obtained:

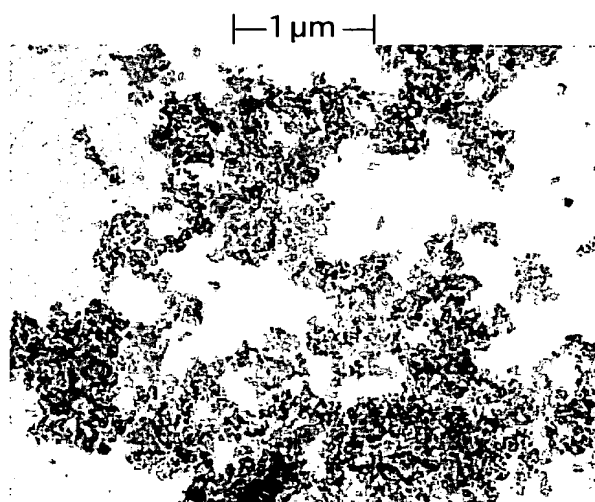


Instant sample B of identical structure was made according to example 1b of the instant application.

The particle sizes were as follows (as observed by transmission electron microscopy):



Comparative sample A



Instant sample B

Comparative sample A consists of large particles essentially of size from about 0.15  $\mu\text{m}$  (few) to 1.0  $\mu\text{m}$  (many).

Instant sample B consists of very small particles essentially of size below 0.1  $\mu\text{m}$ , as instantly claimed.

The CIE 1976 L\*C\*h color space values were measured according to our standard test procedure for PVC. The test samples were made from compositions comprising  $80.0 \pm 0.5$  mg of color pigment, 2.00 g of  $\text{TiO}_2$  (Kronos® 2056), 14.6 g of plasticizer and 26.6 g of PVC (Evipol® SH 7020), which compositions were extruded on a Collin W110E two-roll device at 160°C (6 minutes), then on a Troester-Walze Nr. 5932/65 (20 passes at room temperature / gap  $0.85 \pm 0.05$  mm), then again on a Collin W110E two-roll device at 160°C (1 minute), to obtain  $0.34 \pm 0.01$  mm thick foils. The results were as follows:

Sample	L*	C*	h
A (comparative)	63.5	37.8	6.35
B (instant)	61.0	39.1	4.52

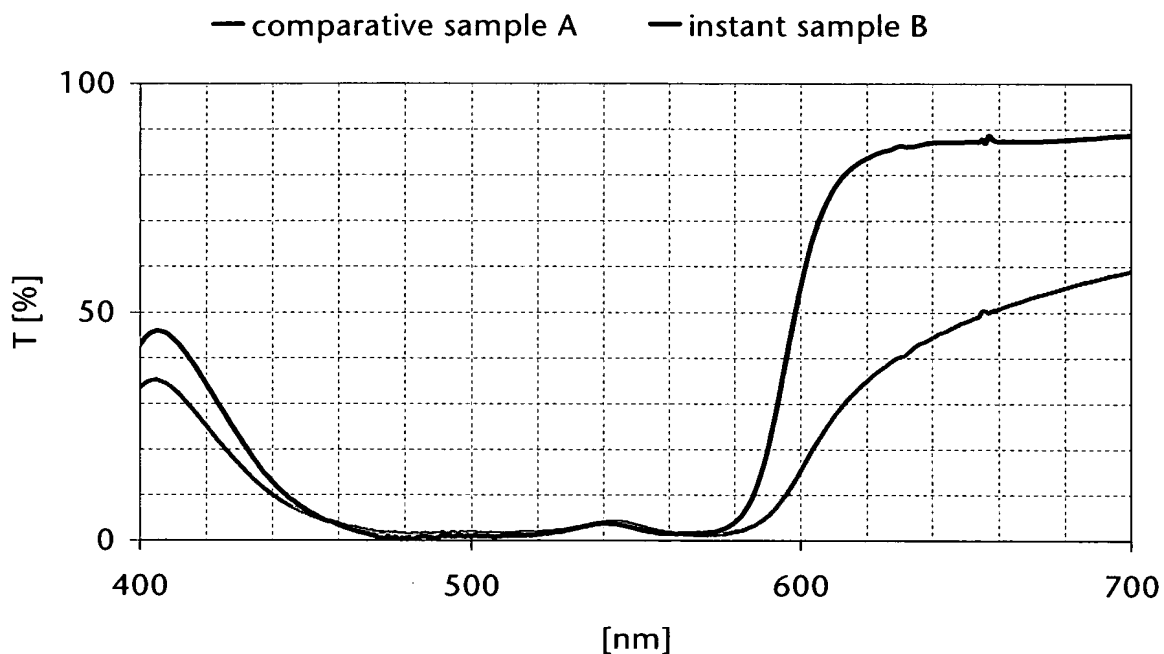
In comparison with comparative sample A made according to the prior art process of US Patent No. 4,579,949 (Rochat et al.), the instant sample B has a higher color strength (lower L\* value), higher color saturation (C\* value) and more bluish hue (h value).

The transmission spectra were further determined, using samples made from compositions comprising  $80.0 \pm 0.5$  mg of color pigment, 14.6 g of plasticizer and 26.6 g of PVC (Evipol® SH 7020), which compositions were extruded as described above, however finally to  $0.21 \pm 0.01$  mm thick foils.

The instant sample B had a transmission of below 5% (1.6-4.7%) at 570-580 nm and above 80% (81.3%) at 615 nm. This matches the requirements of the instant claims. In comparison, comparative sample A had a transmission of much below 80% (only 31.5%) at 615 nm. This does not match the requirements of the instant claims.

Moreover, instant sample B had an extremely higher relative transparency ( $\Delta Tr$ ), as compared with comparative sample A (measured over black according to the test procedure standardly used in our screening labs).

The PVC transmission spectra are shown comparatively on the following graph:



Color filters were further made according to example 7 of the instant application, using the above samples A and B at same concentration. The Yxy color space was used, which is the standard in the field of color filters. The results were as follows:

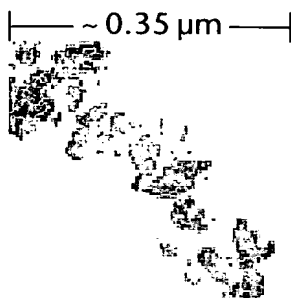
Sample	x	y	Y	contrast ratio
A (comparative)	0.582	0.265	8.11	15
B (instant)	0.579	0.268	14.10	290

In comparison with comparative sample A made according to the prior art process of US Patent No. 4,579,949 (Rochat et al.), the instant sample B has a quite similar color point on the chromaticity diagram (x and y values), however a much higher lightness (tristimulus value Y) and a much higher contrast ratio.

## DISCUSSION

There is no specific example of a pigment of instant structure of formula (1) in '949. Comparative sample A is what the skilled artisan would have obtained if he had phenylmercapto as a substituent ('949 / column 4 / line 25), while otherwise performing the process as disclosed in example 48, which is the nearest to the instant process. Comparative sample A consists of very large particles.

As revealed by a greater magnification (from a higher resolution picture), instant sample B on the contrary consists of highly crystalline, relatively flat particles having an irregular border:



This is much different from particles that are obtainable by conventional pigment comminuting methods such as salt-kneading, which are in general much more spherical due to the influence of high shearing forces and mechanical energy input.

As demonstrated by above experiments, the instant sample B has clearly different coloristic properties, as compared with the comparative sample A. The instant process is also quite different from that of '949 in that the pigment is finally obtained by the single step of discharging the reaction mixture into a mixture comprising methanol and acetic acid at a temperature below 30°C. In contrast, example 48 of '949 discloses the addition of methanol to the 60°C warm reaction mixture, followed in a second step by the addition of a mixture of glacial (= water-free) acetic acid and methanol to the reaction mixture. In this prior art process, crystal seeds are first formed upon dilution, which then grow rapidly to a large size as shown in above electron microscopic pictures. The end temperature in the process of '949 is in any case higher than 30°C, due both to the residual heat of the reaction mixture and apparatus and to the heat of neutralisation.

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The pigments obtained by the instant process have a higher transmission, especially at 615 nm, as well as higher color strength, a more bluish hue and a much higher contrast ratio in color filters, which properties are all highly desirable. Their slope on the bathochromic side of the absorption is advantageously very steep in the range from 580 to 615 nm.

## CONCLUSION

The instant particles show a unique combination of a very tiny particle size having a flat, crystalline shape, which combination leads to surprisingly improved coloristic properties especially useful in color filters.

This advantage could in no way be expected from the prior art and was obtained through clearly unobvious modifications of the general process of manufacture disclosed in '949, both changing the temperature and reversing the way of mixing the reaction mixture and the quenching medium, while further also omitting the dilution step.

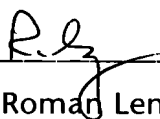
WO-02/10288 does not teach any method for synthesizing diketopyrrolopyrrole pigments. It also fails to disclose any specific example of a diketopyrrolopyrrole pigment having the instant substituents comprising sulfur. Though their particle size is below 0.1  $\mu\text{m}$ , the pigments of '288 are structurally and coloristically different from the instantly claimed pigments.

Furthermore, the combination of '949 with '288 does not lead to my invention. As clearly shown by above experimental evidence, a color filter made using comparative sample A has a much too low transmission at 615 nm.

Hence, US Patent No. 4,579,949 (Rochat et al.) and PCT patent application WO-02/10288 (de Keyzer), either alone or in combination with each other, fail to disclose or suggest the instant invention, which is hence clearly novel and not obvious.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

Signed, this 21<sup>st</sup> day of March, 2007

  
Roman Lenz